PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Arne GODAL et al. Conf. No.: 5309

Appln. No.: 09/674,661 Group Art Unit: 1754

Filed: July 02, 2001 Examiner: Stuart L. HENDRICKSON

For: PROCESS FOR PRODUCING CARBONACEOUS SOLID MATERIAL AND HYDROGEN-RICH GASES

DECLARATION UNDER RULE 132

Assistant Commissioner of Patents
P.O. Box. 1450
Alexandria, Virginia 22313-1450

I, Arne Godal, a Norwegian citizen of Fanaveien 289B, N-5244 Fana, Norway; and

I, Bjorn Magnussen, a Norwegian citizen of Lassvebergvegen 19, N-7562 Hundhamrarn, Norway,

declare as follows:

1. We are inventors on the present application. We have reviewed the Office Action dated 30 November 2004 which issued on the above application, wherein the Examiner raised objections under 35 U.S.C. 103(a), that the claims are obvious over Lynum et al (US 5,527,518), and under 35 U.S.C. 102(b) or in the alternative 35 U.S.C. 103(a), that the claims are anticipated by, or are obvious over, Morgan et al (US 3,619,140). We are informed that the principle issues of concern are that in both cases a flame using oxygen is used followed by a substantially oxygen-free pyrolysis/decomposition step to make the carbon black product which is not different to the claims on file. This Declaration serves to illustrate how the teachings differ from the claimed method.

2. Lynum et al employs a plasma torch for the first stage of

pyrolytic decomposition. This involves the generation of a plasma field by forcing a steady flow of gas between electrodes with a high electrical current flowing between them. The ionized gas generates the intense heat required for pyrolytic decomposition. Lynum et al states in column 5, line 25 that the plasma gas is preferably hydrogen. Thus, Lynum et al does not use a flame using oxygen. If such a flame had been used some combustion would be expected and Lynum et al only refers to pyrolytic decomposition in this first stage.

3. The use of oxygen is only suggested at the second stage of the process. The second stage involves the complete decomposition of the dehydrogenated carbon material formed by the first stage of pyrolytic decomposition (see e.g. claim 1). It is suggested that additional raw materials may be added to quench the already formed carbon black (see column 5, lines 45-48 and claim 1). Such raw material may include oxidants such as oxygen (column 2, line 46-51). The method of Lynum et al is therefore not reliant on the use of oxygen and the method can be used without oxygen. Oxygen is only suggested for post-production modification. We understand that the claims require that the hydrocarbon fuel is heated in the presence of oxygen. However, as mentioned above, in the Lynum et al method, oxygen is added at the second stage to influence the carbon black already produced and is thus not provided in combination with the hydrocarbon fuel unlike in the claimed method.

4. We understand that the claims require that the hydrocarbon fuel undergoes incomplete combustion and partial pyrolytic decomposition. However, since the oxygen which may be added by the method of Lynum et al is added once the carbon black has been formed and is stated to be useful for quenching, no combustion occurs and thus the method of Lynum et al does not involve the required combustion.

5. The product that is produced according to the claimed method is different to the product produced according to Lynum et al. The table in Annex 1 reflects the measurements made on the carbon black produced according to the method of the invention and tested by the

Deutsches Institut fur kautschuktechnologie in Germany. Thus the claimed method yields carbon black with a particle size from 20-30nm and a surface area of approximately $110m^2/g$. In contrast Lynum et al produces particles with a surface area of <5 to $30m^2/g$ (see column 6, lines 1-14). This correlates to a particle size of approximately 80-320nm. The smaller particles generated in accordance with the claimed method are of considerable value in the conductor and pigment industries.

6. The method as claimed is based on a fundamentally different premise to the method of Lynum et al. In the case of Lynum et al, carbon black is created by pyrolytic decomposition. Alteration of the product's properties such as particle size, density and quantity is achieved by adding additional raw materials to cause quenching (see column 5, lines 45-49). In contrast in the claimed method such control is exercised by controlling the amount of oxygen present in the combustion process in view of the crucial role that oxygen plays in the claimed method.

7. The combustion of fuel with controlled amounts of oxygen provides controlled combustion and localized heating leading to spontaneous formation of nuclei which are the precursors to carbon black particles. As the process is strongly endothermic, the temperature in the reactor drops as the carbon black particles are formed until the reactions causing particle formation are quenched. This happens automatically and is achieved by controlling the amount of oxygen in the system and hence avoids combustion of the carbon black that is formed. This introduces an internal element of control into the system and by varying the amount of oxygen and hence combustion, particles of different sizes may be made without the need to introduce any quenching reagents.

8. Thus the claimed method offers several advantages over the Lynum et al method. No additional reagents are required to modify or control particle size which avoids potential contamination by contact with a quenching agent. Particle size may be easily modified by modifying the amounts of the starting materials. Automatic, internal, quenching provides consistency not reliant on a second stage of quenching which may introduce an element of

variability. The claimed method is also highly efficient directly using the energy in fuel rather than relying on the production of electricity to generate a heat source which itself generally requires the burning of a hydrocarbon fuel.

9. Lynum et al thus concerns an entirely different process. Lynum et al does not recognize any role for oxygen in generating carbon black or that this could be used to generate an internal quenching system to do away with the quenching step in the Lynum et al method. The method of Lynum et al is concerned with a method of pyrolytic decomposition and the use of a plasma torch specifically allows the method to be conducted without combustion. The introduction of a combustion step would entirely alter the method as suggested by Lynum et al and is specifically avoided by the use of a plasma flame. We see no reason that the method would be converted to include combustion, particularly since the advantages which could be achieved using such a process are not recognized.

10. Morgan et al makes carbon black based on a combustion method. As stated in column 1, lines 61-63 the method is concerned with processes for improving oxygen-enriched carbon-black processes. Combustion products are generated in a burner by mixing and heating a flame fuel and oxygen and these products are then mixed with a hydrocarbon oil at high temperature to produce carbon black. The reaction is then quenched with water.

11. The method of Morgan et al is carried out with high concentrations of oxygen which significantly exceed the amounts claimed in the present claims. Annex 2 illustrates the C:O ratio used in Morgan et al by reference to Examples 1-5 within that document. It will be noted that the C:O ratios used are 1:1.6 or 1:1.7 and thus reflect an oxygen-rich system. This is in contrast to the claimed method which states that the ratio of C:O is greater than 1:0.4. Thus the Morgan et al method uses 4 times the maximum amount of oxygen used in the claimed method.

12. The high amount of oxygen used in the Morgan et al method is evident in the % combustion that is achieved. The Examples refer to combustion at levels of from 29.1 to 31.2%. This far exceeds

the levels of combustion achieved by the claimed method. As mentioned in paragraph 11, the maximum use of oxygen according to the claims constitutes a 1:0.4 C:O ratio. The highest theoretical possible extent of combustion that can be achieved using such controlled low levels of oxygen is 10% (see the calculation in Annex 3). For the oxygen to be present in non-limiting levels a ratio of 1:4 would need to attained. Morgan et al uses C:O at a ratio of 1:1.6 or 1:1.7 (see Annex 2). With oxygen at this level a theoretical maximum of 40-42.5% combustion could be achieved (see Annex 3). The method of Morgan et al approaches this maximum.

13. Morgan et al provides no evidence that the amount of oxygen should be modified or that in doing so advantages would be achieved. Morgan et al uses a quenching step and provides no teaching that this could be avoided by modifying the amounts of oxygen used in the system.

14. Thus the methods of Lynum et al and Morgan et al may be clearly distinguished from the claimed method. In the case of Lynum et al oxygen is not used to achieve partial combustion. In the case of Morgan et al, the amount of oxygen used is well outside the claimed maximum levels of oxygen that may be used. In neither case is there motivation to remedy these deficiencies since they are concerned with distinct methods of making carbon black neither of which are concerned with limited combustion to produce a self-quenching pyrolytic decomposition reaction.

15. We further declare that all statements made herein of our own knowledge are true, and that all statements made on information and belief are believed to be true, and that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Codes, and that such wilful false statements may jeopardize the validity of the application and any patent issuing thereon.

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Arne Godal Date

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Bjorn Magnussen Date

ANNEX 1

EXPERIMENTAL PARAMETERS OF CARBON BLACK PRODUCED ACCORDING TO THE CLAIMED METHOD

Carbon black was generated according to the claimed method as described in the patent application in issue.

Carbon black with a particle size of from 20-30nm (average 24nm) was generated. Surface area measurements were made according to two different methods and the results are shown below.

Test method	Description of test	Surface area
ASTM D1510	Iodine Absorption number	112.4±0.7mg/g
ASTM 3037	Surface area by Nitrogen adsorption (NSA)	107.5m^2/g

ANNEX 2

CALCULATION TO DERIVE THE C:O RATIO IN THE EXAMPLES OF US 3,619,140

Example 1 uses a combustion step mixing together 1,600 cubic feet per hour oxygen with 800 cubic feet per hour natural gas in a burner. The combustion products (and unreacted products) are then passed into a carbon-forming zone together with carbon-black make oil at a rate of 12.5 gallons per hour.

In the calculation below, some approximations have been made for ease of calculation or because relevant information is absent. These should however only affect the results to a minor extent.

EXAMPLES 1-3

Reagents introduced into the system in 1 hour:

1,600 cubic feet oxygen
800 cubic feet natural gas
12.5 gallons carbon-black make oil

For ease of calculation, natural gas has been assumed to be 100% methane. American natural gas comprises 85% methane, the remainder being made up of ethane, propane, nitrogen and butane. A ratio of 1C:4H does however provide a close approximation to the composition of natural gas. The type of carbon-black make oil is not stated. However, again for ease of calculation, the simplest hydrocarbon oil has been used, liquid methane. Whilst this may not be practical for the Morgan et al method, it is used here for calculation purposes. The above amounts may be converted into stoichiometric values as follows:

Oxygen
O_2 gas has a density of 1.429g/l. This equates to 0.089 moles of oxygen atoms/l. Oxygen was provided at 1,600 cubic feet per hour which equates to 44,880 l per hour (1 cubic foot = 28 l). 3994 moles of oxygen were therefore provided per hour.

Natural gas

Methane gas has a density of 0.7168g/l. This equates to 0.045 moles methane (or carbon)/l. Methane gas was provided at 800 cubic feet per hour which equates to 22,440 l per hour (1 cubic foot = 28 l). 1010 moles of methane (and hence carbon) were therefore provided per hour.

Make oil (liquid methane)

Methane liquid has a density of 422g/l. This equates to 26.38 moles methane (or carbon)/l. Make oil was provided at 12.5 gallons per hour which equates to 56.88 l per hour (1 gallon = 4.55 l). 1500 moles of methane (and hence carbon) were therefore provided per hour.

Total moles of carbon per hour: 1010 + 1500 = 2510
Total moles of oxygen per hour: 3994

Ratio C:O - 1:1.6

EXAMPLES 4 AND 5

Relative to Examples 1-3, Examples 4 and 5 use a quarter of the natural gas, but increase the make oil from 12.5 to 16.7 gallons per hour. Oxygen remains the same.

Total moles of carbon per hour: 253 + 2004 = 2257
Total moles of oxygen per hour: 3994

Ratio C:O - 1:1.7

ANNEX 3

EFFECT OF OXYGEN LEVELS ON EXTENT OF COMBUSTION

Complete combustion using methane as the carbon source and O_2 as the oxidant may be represented as:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$$

In this equation, the C:O ratio of the starting materials is 1:4.

If the oxygen levels are limited such that the C:O ratio is 1:0.4, one tenth of the level required to achieve complete combustion is provided and hence the maximum theoretical combustion that may be achieved is 10%.

A C:O ratio of 1:1.6 or 1:1.7 (as used by Morgan et al) provides 40 or 42.5% of the required oxygen for complete combustion, thus allowing a maximum of 40 or 42.5% combustion.